



Mode resolved density of atmospheric aerosol particles

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J. Kannosto et al.

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

In this study, we investigate the mode resolved density of ultrafine atmospheric particles measured in boreal forest environment. The method used here enables us to find the distinct density information for each mode in atmospheric fine particle population: the density values for nucleation, Aitken, and accumulation mode particles are presented. The experimental data was gained during 2 May 2005–19 May 2005 at the boreal forest measurement station “SMEAR II” in Hyytiälä, southern Finland. The density values for accumulation mode varied from 1.1 to 2 g/cm³ (average 1.5 g/cm³) and for Aitken mode from 0.4 to 2 g/cm³ (average 0.97 g/cm³). As an overall trend during the two weeks campaign, the density value of Aitken mode was seen to gradually increase. With the present method, the time dependent behaviour of the particle density can be investigated in time scale of 10 min. This allows us to follow the density evolution of the nucleation mode particles during the particle growth process following the nucleation burst. The density of nucleation mode particles decreased during the growth process. The density values for 15 nm particles were 1.2–1.5 g/cm³ and for grown 30 nm particles 0.5–1 g/cm³. These values are consistent with the present knowledge that the condensing species are semi volatile organics, emitted from the Boreal forest.

1 Introduction

Atmospheric aerosol contains a complex mixture of particles of natural and anthropogenic origin, consisting of both primary and secondary particles from a variety of sources (e.g., Seinfeld and Pandis, 1998). Its chemical and elemental composition, optical, hygroscopic and other important physical and chemical properties vary both spatially and temporally. Additionally, there is variation of all these quantities over particle size. The assessment of climatic and adverse health effects of atmospheric aerosol particles requires detailed information on particle properties (Yu et al., 2006; Lohmann

ACPD

8, 7263–7288, 2008

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

and Feichter, 2005; Pope and Dockery, 2006). For example, the particle size and chemical composition affect the particle residence time in the atmosphere, its optical properties and its potential to act as cloud condensation nuclei as well as its behaviour in human lung. In addition, the particle chemical composition and other chemical and physical properties carry information concerning sources and formation mechanisms of particles.

Atmospheric fine particles can be classified to ultrafine ($d_p < 0.1 \mu\text{m}$) and accumulation mode ($0.1 \mu\text{m} < d_p < 1 \mu\text{m}$) particles. Ultrafine particles can be further divided into nucleation ($< 30 \text{ nm}$) and Aitken mode particles ($30 \text{ nm} < d_p < 0.1 \mu\text{m}$). The ultrafine size range dominates the particle number concentration, and in urban areas usually contributes to more than 90% of particle total number. Generally, after their formation, the fresh aerosol particles in atmosphere appear in this size range. In addition, being also in the critical size range for acting as cloud condensation nuclei, ultrafine size range affects the number concentration of cloud droplets in remote and moderately polluted environments.

The composition and properties of accumulation mode particles are fairly well known, because proper measurement and analytical methods have been available for this size range and mass concentration level (e.g., Wall et al., 1988; Seinfeld and Pandis, 1998; Maenhaut et al. 1999; Putaud et al., 2004). A few studies in distinct locations have provided also information on the composition of the ultrafine size mode, such as existence of polycyclic aromatics in urban aerosol (Allen et al., 1998), overall urban aerosol at around 100 nm (Cass et al., 2000), elemental composition of chlorine in sub 10 nm coastal aerosol (Mäkelä et al., 2002), distinct chemical components such as amines in boreal forest (Mäkelä, 2001a), organics vs. in-organics in boreal forest (O'Dowd et al., 2002) and organics in coastal aerosol (Vaattovaara et al., 2006). Recent results obtained by mass spectrometers give first insights to the full chemical composition of ultrafine particles (Zhang et al., 2005; Smith et al., 2005; Allan et al., 2006). However, there are still clear gaps in the overall knowledge about the ultrafine size range.

Nucleation bursts observed in different environments are producing nucleation mode

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

particles in atmosphere (Kulmala et al., 2004a). There have been intensive and successful efforts to identify different nucleation mechanisms (e.g. Kulmala et al., 2006). Regardless of the progress, there are still gaps in the general understanding of the new particle formation, and between the experimental data and models. In the experimental data there are for example long time series available showing particle size evolutions after nucleation event (e.g., Dal Maso et al., 2005), but the observations of composition and properties are mostly lacking. It is not clear which compounds are responsible for condensational growth process of nucleation mode particles (e.g., Kulmala et al., 2004b). A number of different organic species has been linked with the particle growth process in natural forest environment (e.g., O'Dowd, 2002; Tarvainen et al., 2005; Laaksonen et al., 2007)

As a property complementary to particle composition, the density of the particles is of great interest. Accumulation and coarse mode particle density has been studied e.g. by McMurry et al. (2002) and Saarikoski et al. (2005). In this study, we investigate mode resolved density of atmospheric ultrafine particles in boreal forest environment, using a density fitting method (e.g., Virtanen et al., 2006). Preliminary, time averaged density values have been discussed by Kannosto et al. (2006). Here we present analysis of the full data set, reporting values for nucleation, Aitken, and accumulation modes. As a new aspect, we study the time dependent behaviour of the particle density. This allows us to follow the density change of particles during the particle growth process following a nucleation burst.

2 Method description

Particle density was estimated using the parallel measurement method developed at Tampere University of Technology, first described by Ristimäki et al. (2002). The method is based on simultaneous ("parallel") distribution measurement with ELPI and SMPS/DMPS and further on the relationship between particle aerodynamic size, mobility size and effective density. First applied to laboratory aerosols and diesel exhaust

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



particles, the method has been developed further to be suitable for atmospheric aerosol (Virtanen et al., 2006; Kannosto et al. 2006). We have now further modified the data treatment method to be suitable for large data series and multimodal distributions which are typical for Boreal forest environment.

As the shape of the aerosol distributions are commonly relatively close to lognormal distributions, the measured number distribution on mobility axis is divided into log-normal sub-distributions (modes). The automatic algorithm described by Hussein et al. (2005) is used in fitting the lognormal distributions to measured DMPS or SMPS data. In order to find the correct density for each mode the ELPI response to the multi-modal distribution is simulated. In the calculation of the simulated ELPI currents a mathematical model of ELPI is used. The complete set of information, required to construct the mathematical model of ELPI, can be found in Marjamäki et al. (2005).

In the calculation of the simulated currents, the effective density of particles is needed. By changing the effective densities of modes the simulated ELPI currents are changed. The simulated ELPI currents are obtained by summing up the currents produced by each sub-distribution. The main difference to the single mode case (Ristimäki et al., 2002) is that instead of one effective density, effective density for each mode is searched. This means that the search algorithm has to operate in multi-dimensional space and result may be more sensitive to the start point than with single mode case. Several methods can be used to define the start point for the search algorithm. We have initialized the effective densities with an *ad hoc* method where the initial effective densities are suggested in a sequence starting from the distribution contributing most of the ELPI current and ending with the one that contributes least. These contributions can be easily calculated as the total current produced by distribution does not depend on the effective density. The actual initialisation of each mode is performed by utilizing a set of pre-selected densities ranging from 0.1 to 10 g/cm³, and defining the density which gives the smallest difference between measured and simulated currents. After the initialisation the search algorithm (the “fminsearch” function of Matlab®) minimises the difference between the measured and simulated ELPI currents by changing the

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

effective densities of each mode.

2.1 Method evaluation

The method used in this article has been previously evaluated in laboratory setting by Virtanen et al. (2006). They used laboratory aerosols with bimodal size distributions.

5 The particles consisted of Fomblin (perfluorinated polyether inert fluid, Ausimont Ltd, $\rho=1.9\text{ g/cm}^3$) and di-octyl sebacate (DOS, $\rho=0.91\text{ g/cm}^3$) and the geometric mean diameters of two modes were varied between 40–150 nm. The method produced density estimates of $(0.8\pm0.08)\text{ g/cm}^3$ and $(1.8\pm0.26)\text{ g/cm}^3$ for DOS and Fomblin respectively. Some additional laboratory tests with DOS and smaller particle sizes were performed
10 by Ristimäki (2006). In Fig. 1, the results measured with carefully calibrated impactor are shown (black dots in Fig. 1). The lowest mode size measured was 13 nm. The agreement with bulk value of DOS (gray line in Fig. 1) and measurement results is good. In the Fig. 1 also the earlier laboratory test results reported by Ristimäki et al. (2002) are shown (open circles). Those points are calculated by using the older
15 impactor kernel functions, which had some inaccuracy in the cut of curve of the 1st impactor stage. The figure clearly shows that a careful calibration of the impactor is essential.

Because of the number of variables affecting the performance of the method, laboratory tests are not an efficient way to find out the restrictions of the density fitting
20 method. Thus a large number of simulations were made to find out the “detection limit” of the method. The simulations show that the detection limit depends mainly on GMD of modes and relative concentration of modes. According to the simulations, the method is usable and occasional wrong results are not statistically significant. According to simulation runs, the density analyzing method is capable of evaluating density
25 of nucleation mode particles in multimodal distribution, even when the smallest mode GMD is around 15 nm. The density results of nucleation mode particles in multimodal distributions can be accurate if circumstances are favourable. First of all, this requires sufficiently high concentrations to get a good mode fitting result from the SMPS/DMPS

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



measurement. In addition, the nucleation mode must generate enough current in the ELPI in order for our method to arrive at a reliable density estimate. If approximately 20% of the measured current is produced by the nucleation mode the density value can be found.

5 According to method runs made with simulated data, there are a few situations when the method has an increased possibility to yield incorrect density results. These situations are encountered in cases where two modes both generate approximately the same amount of ELPI current or if the current generated by the one mode is insignificant. In the former case, when generating the ELPI response, the two modes may
10 swap places on the aerodynamic axis ("mode swapping"). This can be recognized as the other mode receives an improbably high density value, while the other one receives an improbably low one. In the latter case, the mode producing the small ELPI current may receive almost any density value without affecting the density of the dominant mode significantly. Both types of error can be recognized and the erroneous density
15 values removed during analysis.

The sensitivity of the particle density calculation to measurement errors was investigated using the method first employed by Virtanen et al. (2004). In order to see how easily the particle density results are affected by measurement error, we took the existing measurement data and added a random, normally distributed 5% noise component to each of the measured ELPI current channels. For each measurement point,
20 the density analysis was then run with 100 noisy samples. In most cases, the standard deviation of the density result was less than 10%. We used high standard deviation values to remove the less reliable data points from further analysis. The measurement points where the added noise caused mode swapping received high standard deviation
25 and the points were removed. The data points with one dominant mode tended to receive high standard deviation values for the less dominant mode density. The density data for the less dominant mode were removed from analysis. As the more dominant mode density values did not receive high standard deviation, they were not removed.

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

3 Description of measurement campaign

The experimental data was gained during 2 May 2005–19 May 2005 at the boreal forest measurement station “SMEAR II” in Hyytiälä (61°51′N, 24°17′E, 181 m a.s.l.), southern Finland. Hyytiälä is background measurement station at southern Finland and around the SMEAR there are extended areas of Scots Pine (*Pinus sylvestris*). Concentrations are typical background levels except wind directions 215°–265° where city of Tampere and station buildings are located. During the 16 (from noon) and 18 (until the morning) May the instruments were not in use. The measurement site has been presented more detail in Kulmala et al. (2001) and by Hari and Kulmala (2005).

The particles size distributions were measured with four separate instruments: ELPI (Dekati Ltd), outdoor ELPI (Dekati Ltd), SMPS (CPC 3025, DMA 3071, TSI), and DMPS two Vienna type DMAs, with 11 cm and 28 cm tube lengths, CPC 3025, CPC 3010 (Mäkelä et al., 1997). The measurement range of ELPI (with 10 lpm flow) and outdoor ELPI (with 30 lpm flow) were 7 nm–6 μ m and measurement time resolution was 1 min. The SMPS size range was ab. 10 nm–400 nm and time resolution 10 min. The DMPS data is a combination of two parallel DMPS and the overall measurement size range was range ab. 3 nm–500 nm. Measurement instruments located in two different cottages (ELPI and SMPS in one and outdoor ELPI and DMPS in another) and distance between the two cottages was about 70 m. The SMPS and ELPI sampling line was \varnothing 12 mm steel tube and the inlet height was 4 m. The outdoor ELPI and DMPS had own inlets and sampling lines. Inlets were at 4 m high and sampling lines were \varnothing 8 mm steel tube. The results shown in this paper are measured by using DMPS and ELPI (10 lpm) unless otherwise noted.

The weather during the measurements was typical for Finnish spring. Temperature varied between 1 to 22°C and average temperature during daytime was 10°C and during night time 5°C. First days (2–8 May 2005) wind direction was between South-West-South-East (100°–217°) and during 8–14 May 2005 wind direction turned towards north (300°–50° most of the time) and last days (14–19 May 2005) wind direction was mainly

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

150°–260°. There was some rain 3 May 2005 and 5 May 2005 evening and night, 15 May 2005 day and night and during the night 16–17 May 2005. In Fig. 2 the particle concentrations measured during the campaign are presented as a contour plot (Fig. 2a) and as a time series (Fig. 2b). The particle formation episodes were recorded during 3, 8, 11–15 May and 17–18 2005.

4 Results

In Fig. 3a, all density values gained during the campaign are plotted as a function of mode GMD. The atmospheric particles are typically grouped into different modes according to mode GMD (Mäkelä et al., 2001b). In our data treatment we have also grouped the results according to mode diameter. All GMD values presented in this paper are mobility equivalent sizes unless otherwise mentioned. As can be seen, the density of nucleation mode particles (15–30 nm) varies between 0.5 and 1.5 g/cm³. Let us next investigate the density results of the Aitken and accumulation mode particles. For Aitken mode particles we have chosen the size range 30–70 nm instead of the often used 30–100 nm (e.g., Hussein et al., 2004; Koponen et al., 2003). The reason for the chosen size range is justified when Fig. 3a is investigated in more detail. It shows that the particle density values are separated into two groups, depending on their mode GMD. Particles with the mobility based mode diameter smaller than 70 nm clearly have lower density values than the larger particles. The separation of atmospheric aerosol particles into different modes is based on the assumption that the modes have different origin (e.g., Seinfeld and Pandis, 1998). Thus it can be assumed that the modes have also different compositions and density. The clearly distinguishable density values of particles in the two groups shown in Fig. 3 support this assumption. According to these results, we can conclude that during our measurement campaign in Hyytiälä the justified size limit (mobility based) for Aitken and accumulation modes was around 70 nm. In Fig. 3b, particle density is presented as a function of the aerodynamic particle diameter. The aerodynamic diameter is related to particle mass and therefore also

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

to particle density. In Fig. 3b it can be seen that the modes are even more clearly distinguishable on the aerodynamic size axis than on the mobility axis. The aerodynamic size limit for Aitken and accumulation mode is approximately 100 nm during our campaign in Hyytiälä. It seems that the particle density measurement gives valuable information that can be used when the particles are to be separated into Aitken and accumulation modes. Such a clear difference in density values of Aitken mode and nucleation mode particles was not detected.

The density results reported in this study are also compared to values reported by Saarikoski et al. (2006) (Fig. 3a and b). They studied the chemical composition of particles collected with a low pressure impactor in Hyytiälä. The density was derived from the mass closure analysis. The agreement of the results presented by Saarikoski et al. and the results presented in this paper is good. During the campaign the size distributions were measured with SMPS and DMPS and also by ELPI (10 lpm) and outdoor ELPI (30 lpm) as mentioned in the previous section. The densities gained by using different instrument pairs were in good agreement: the difference was 17% at maximum. This confirms that the density analyzing method can be applied to different instrument pairs having different characteristics if the instruments are carefully calibrated as was the case here.

The particle density results plotted at time axis are shown in Fig. 4, where the density values of the Nucleation mode particles (black dots in Fig. 4) with $15\text{ nm} < \text{GMD} < 30\text{ nm}$, Aitken ($\text{GMD } 30\text{--}70\text{ nm}$, open triangles in Fig. 4) and accumulation ($\text{GMD} > 70\text{ nm}$, open circles in Fig. 4) modes are presented. From the evening of 16 May 2005 to morning of 18 May 2005 the instruments were not in use, thus the corresponding data points are missing in Fig. 4. As shown in Fig. 4, the density values of Aitken mode varied from 0.4 to 2 g/cm^3 (average 0.97 g/cm^3) and the values for accumulation mode varied from 1.1 to 2 g/cm^3 (average 1.5 g/cm^3). The found density values for accumulation mode agreed well with those reported in other studies (McMurry et al., 2002; Virtanen et al., 2006; Saarikoski et al., 2005; Cozic et al., 2007). The Aitken and accumulation mode particle densities had lowest values in the beginning of the campaign. As an

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

overall trend during the two weeks campaign, the density value of Aitken mode was seen to gradually increase. During the particle formation episodes that took place at 11–15 May, the concentration of the accumulation mode particles was too low for the successful density analysis. The density values of Aitken mode increased during the campaign and reached their maximum value at 14 of May. The highest values of Aitken mode densities ($0.9\text{--}1.3\text{ g/cm}^3$) were recorded in the period during which the particle formation events took place. Increasing density of Aitken mode particles coincided with decreasing relative humidity and increasing solar radiation, accompanied with a turn in the wind direction towards north. However, a larger amount of data is needed before the dependence of the particle density on the different (meteorological) parameters can be properly analyzed.

The density values for the nucleation mode particles were obtained only when the particle concentration was adequate and the mode diameter equal or larger than 15 nm. According to the simulations, the nucleation mode particle concentration was sufficient for density analysis if the nucleation mode particles produce 20% of the current measured in ELPI. This requirement was fulfilled during the particle formation events. During the events the concentration levels of accumulation mode particles were too low to get reliable mode fitting. Hence only density result of Aitken and nucleation modes are presented. During the night of 15 and 16 May, nucleation mode was present, but a clear increase in particle size was not detected. The density of nucleation mode particles varied between 1.5 and 0.5 g/cm^3 . The density of growing nucleation mode particles was lower during the 11–12 May than during the 13–14 May. This follows the overall density progress of Aitken mode particles during those days. The nucleation mode particles appearing during the nights of 15 and 16 May had generally lower density values than particles appearing during the daytime. On 16 May the particle formation event took place in the afternoon when our measurement devices were not in use.

To investigate the trends in particle density in more detail, two days were chosen, different in particle dynamics. 4 May was a day with practically no nucleation mode

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

present whereas on 13 May there was intense particle formation and a high nucleation mode concentration in the afternoon. The weather information and some gas component concentrations of the two days are presented in Table 1. In Fig. 6c–d, the mode densities are plotted as functions of time. The corresponding GMD sizes are shown in Fig. 6a and b. It should be noted here that in Fig. 6a–b only the GMD values of the modes for which the density analysis was successful are shown. The error bars show the standard deviation estimated with the method described in the Sect. 2 of this paper.

During the 4 May (Fig. 5a), the particle population was stable and no remarkable changes in GMDs and concentrations of modes occurred. On 4 May also the particle density values were stable: the density values for Aitken mode were very low: $(0.5 \pm 0.2) \text{ g/cm}^3$ (Fig. 6c). The accumulation mode densities were approximately $1.4 \pm 0.2 \text{ g/cm}^3$ during the whole day (Fig. 6c). The 4 May represents the first days of the campaign during which the particle density were low.

On 13 May (Fig. 6b) the particle formation event took place at 12 a.m. During the particle formation event and the growth process the nucleation mode concentration was very high and the accumulation mode concentration was very low. The new particles achieved a stable mode diameter of approximately 33 nm at around 6 p.m. The density of the nucleation mode peaking at 17 nm was approximately 1.3 g/cm^3 (Fig. 6b and d). The particle density decreased simultaneously with the particle growth process indicating condensation of some lighter compounds. The particles grew until they reached 33 nm mode diameter, when the density levelled at 1.0 g/cm^3 (Fig. 6d). Starting at 19:20 the mode diameter decreased from 33 nm to 20 nm (Fig. 6b). At the same time, the particle density increased from 1 g/cm^3 to 1.2 g/cm^3 (Fig. 6d).

The density decrease associated with the particle growth process during the particle formation event on 13 May is indicated in Fig. 6 by the dashed arrows. The density values of the nucleation mode particles during this process are presented as a function of mode GMD in Fig. 7. By assuming spherical particles the density of condensing species can be estimated by calculating the change in particle mass during the growth process. In calculations we have assumed that the change in mode GSD corresponds

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

to the increase in the particle size and thus the GSD values and related density values have been used (see Fig. 7). The resulted density estimate for the condensing species was $1.0 \pm 0.2 \text{ g/cm}^3$. The above analysis for the growth process of nucleation mode particles was made also for the events that took place on 3, 11, 12 May. The density data for 14 and 15 May were too unstable for such analyze. On 3 May the density of species condensing on to the nucleation mode particles was $0.5 \pm 0.2 \text{ g/cm}^3$. This is in a good agreement with the low density values of Aitken mode particles detected in the first days of the campaign. On 11 and 12 May the corresponding density was $0.7 \pm 0.3 \text{ g/cm}^3$.

5 Conclusions

We have presented the simultaneously measured density values of atmospheric nucleation, Aitken and accumulation mode particles. The Aitken and accumulation mode particle densities had lowest values in the beginning of the campaign. The density values of Aitken mode increased during the campaign and reached their maximum value at 14 of May. The highest values of Aitken mode densities ($0.9\text{--}1.3 \text{ g/cm}^3$) were recorded in the period during which the particle formation events took place. We could not find any unambiguous dependence of the density changes on individual weather parameters. Increasing particle density coincident with decreasing relative humidity and increased solar radiation, accompanied with a turn in the wind direction towards north. However, a larger amount of data is needed before the dependence of the particle density on the different (meteorological) parameters can be properly analyzed.

Density value of nucleation mode particles decreased during the particle formation events. The density values for 15 nm particles were $1.2\text{--}1.5 \text{ g/cm}^3$ and for grown $\sim 30 \text{ nm}$ particles $0.5\text{--}1 \text{ g/cm}^3$. The density of the species condensing on the particles during the growth process was estimated from the data. The resulted estimates varied between 0.5 and 1 g/cm^3 . According to authors' knowledge there are no prior data on the development of the particle density of nucleation mode during the growth process.

Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

A number of different organic species has been linked with the particle growth process in natural forest environment (e.g., O'Dowd, 2002; Tarvainen et al., 2005; Laaksonen et al., 2007) and the density of those species varies from 0.5 g/cm^3 to 1.9 g/cm^3 . Longer data collection, more detailed analysis and additional measurements of e.g. VOC emissions are needed before the found density can be related to any specific species. This is a subject for further studies.

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J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Mode resolved density of atmospheric aerosol particles

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Mode resolved
density of
atmospheric aerosol
particles

J. Kannosto et al.

Table 1. Weather parameters on 4 and 13 May.

	T	potential T	RH	H ₂ O	wind direction	UV-A	UV-A, during daytime	UV-B	UV-B, during daytime	Net Radiation	Net Radiation	Visibility	NO _x	NO	O ₃	CO ₂	SO ₂
units	°C	°C	%	ppth	°	W/m ²	W/m ²	W/m ²	W/m ²	W/m ²	W/m ²	km	ppb	ppb	ppb	ppm	ppb
4.5.2005	5.9	−0.006	86.7	7.9	115.0	5.1	9.9	0.21	0.43	36.0	93.9	26.7	1.61	0.013	42.3	387.0	0.10
13.5.2005	10.4	−0.004	38.9	4.6	196.2	14.69	28.9	0.57	1.2	175.4	417.1	49.7	0.73	0.0004	45.3	384.7	0.08

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

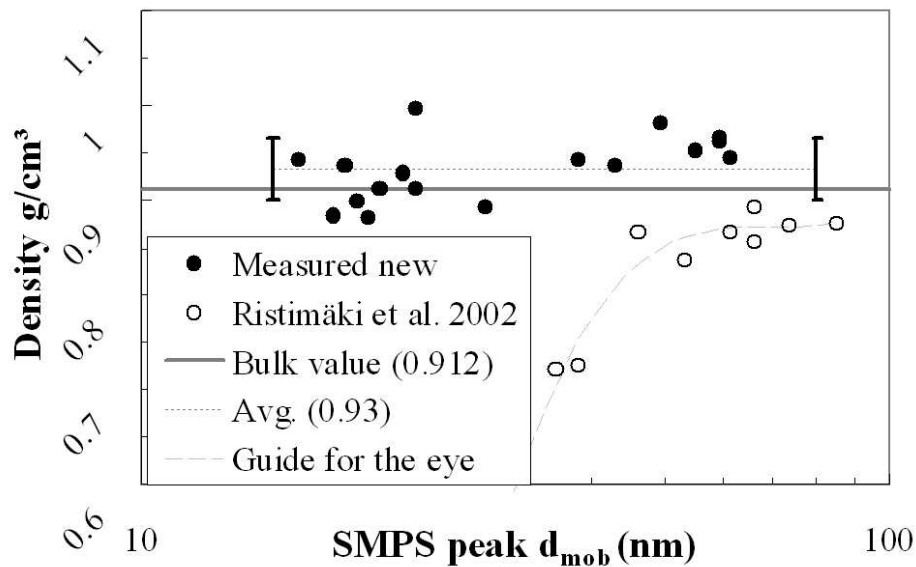


Fig. 1. Laboratory tests of the method using DOS particles. The results gained by carefully calibrated impactor kernels (black dots) are compared to inaccurate kernels (open circles). Figure adapted from Ristimäki (2006).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

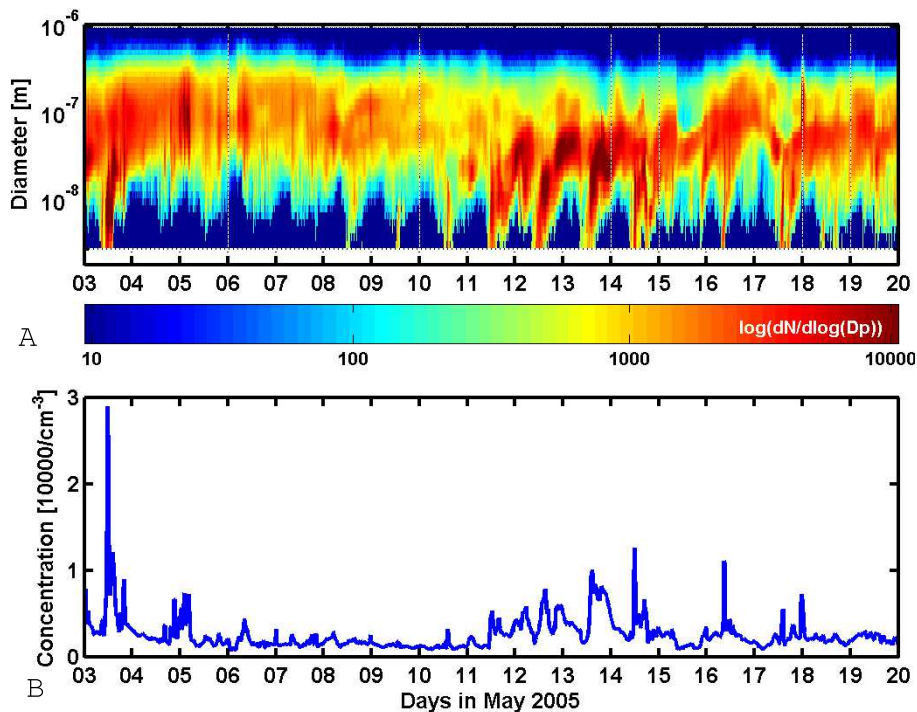


Fig. 2. The number size distributions and total number concentrations measured during the campaign. The concentrations are presented as a contour plot (Fig. 2a) and as a time series (Fig. 2b).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

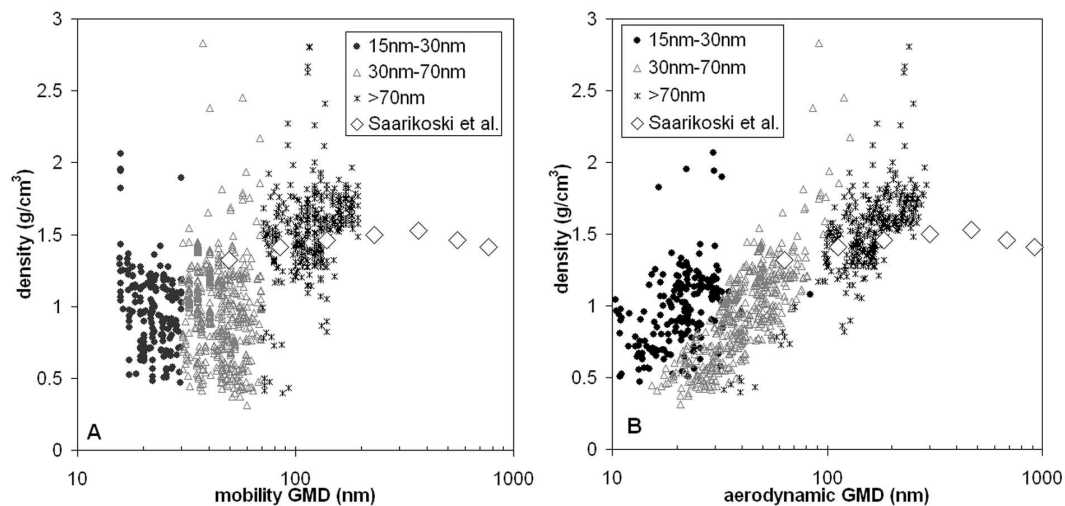


Fig. 3. Particle densities as a function of mobility size **(a)** and aerodynamic size **(b)** of the mode GMD. The results of Saarikoski et al. (2005) are also presented (open diamonds).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

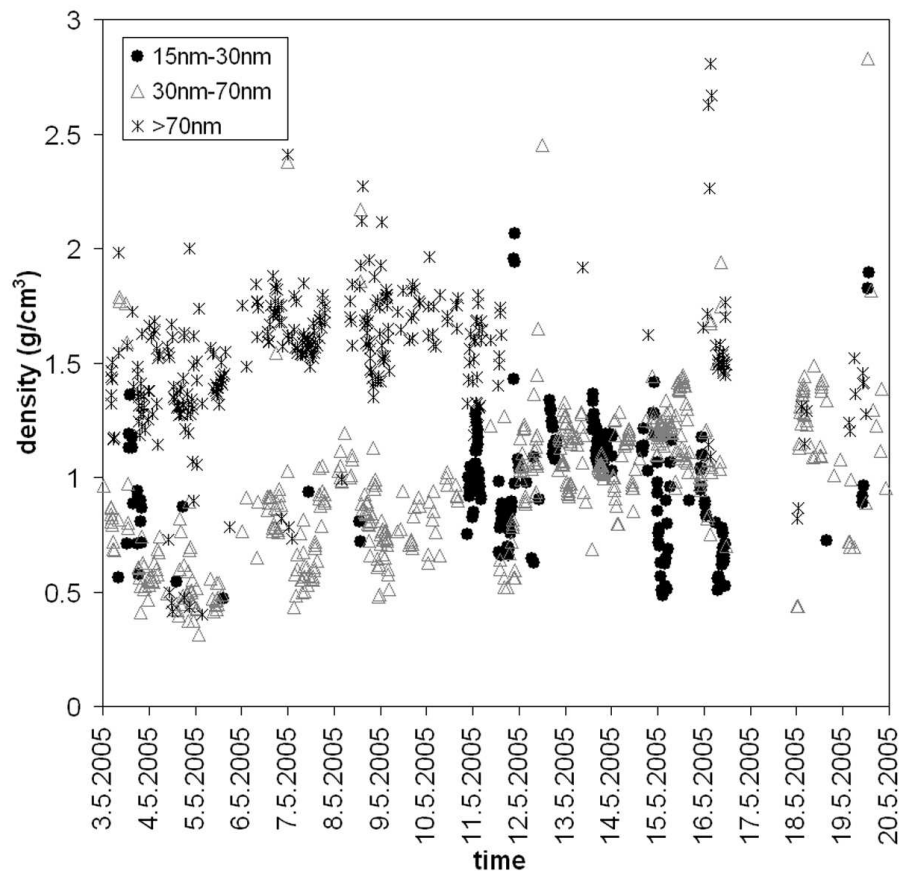


Fig. 4. Particle densities presented as a function of time.

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

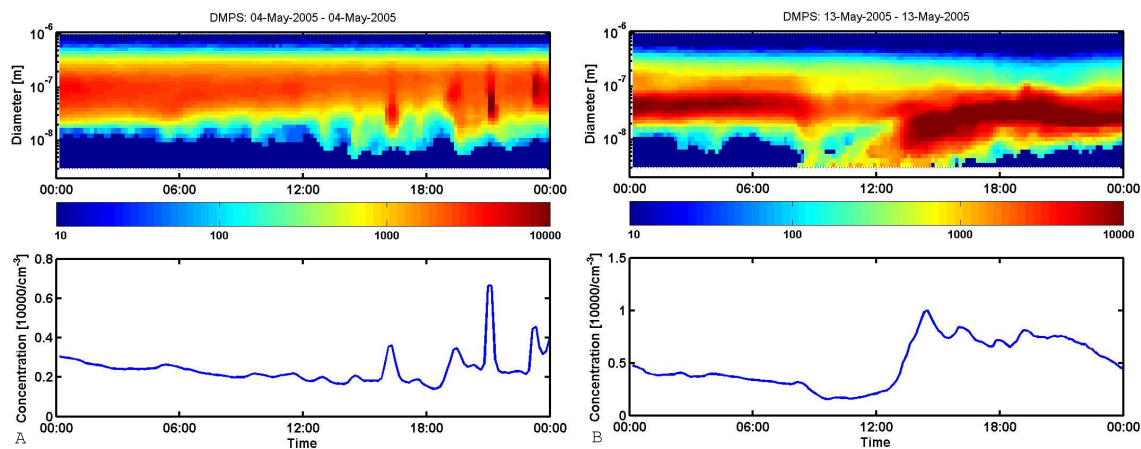


Fig. 5. Particle concentrations and size evolution on 4 May (a) and 13 May (b).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

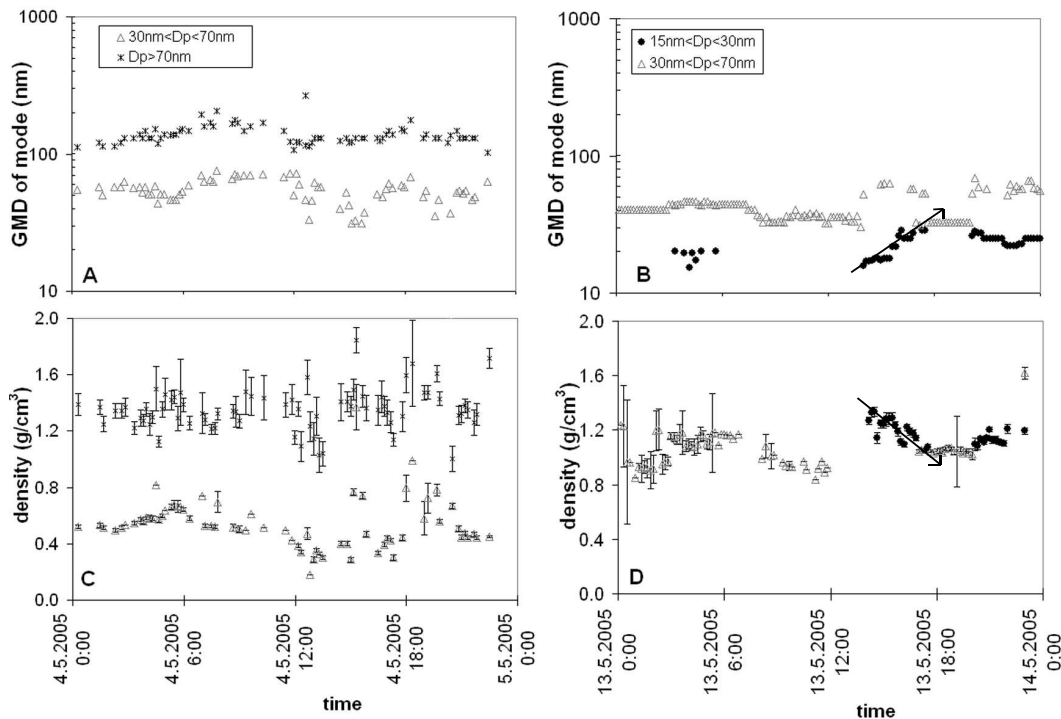


Fig. 6. The densities of separate modes measured on 4 May (a) and 13 May (b), and mode GMDs measured on 4 May (c) and 13 May (d). The dashed arrows show the trend of decreasing nucleation mode particle density during the growth process.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**Mode resolved
density of
atmospheric aerosol
particles**

J. Kannosto et al.

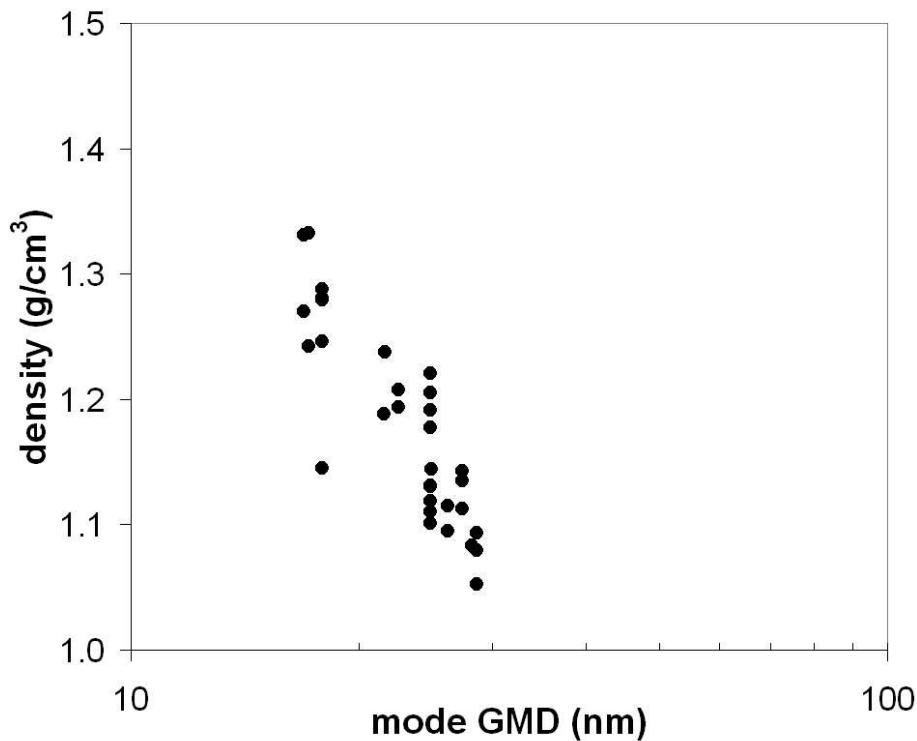


Fig. 7. The density of nucleation mode particles as a function of mode GMD during the growth process on 14 May.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)